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SOLID STATE RESEARCH OF THE APPLIED PHYSICS DEPARTMENT FOR THE YEAR 1965

29 JUNE 1966

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29 June 1966

The Applied Physics Department reports its accomplishments in the fields of solid state, metallurgy and infrared for the year 1965. The support for the work was obtained from Foundational Research and project funds provided by the U. S. Bureau of Naval Weapons, the Bureau of Ships, and the National Aeronautical and Space Administration.

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Captain, USN
Commander

W. W. Scanlon

W. W. SCANLON
By direction

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INTRODUCTION

The research programs in the Applied Physics Department during 1965 cover a number of subjects, ranging from basic research in magnetism and semiconductors to applied research in infrared and metallurgy. We have also contributed directly to the defense effort in Vietnam by conceiving, designing and developing a prototype of an ordnance locator which will be used for searching junks and other vessels in the waters around Vietnam for contraband weapons. Contracts have been let for the production of this device. Ordinarily, we in research, do not pursue device development this far, but in this instance the urgency of the problem required that it be carried through as quickly as possible. The fact that this was all done within a year indicates a certain degree of flexibility in the capability of our personnel.

Semiconductor research continues to be concentrated on small band gap materials which are of interest as infrared detectors, sources, or filters. More recently we have extended this work to still smaller energy gap materials that border on metallic behavior. One of these materials, SnTe, was thought for a number of years to be a semimetal. As a result of the techniques we have developed for making thin single crystal films by epitaxial vacuum evaporation, we were able to measure the optical absorption edge in SnTe and to establish that it is a semiconductor rather than a semimetal.

Epitaxial film techniques are particularly useful in studying alloy compositions. We have been able to prepare alloys of the semiconductor $PbTe_xSe_{1-x}$ over the entire range of x which permits study of the transition in band structure as a function of composition.

Our continued interest in surface effects in metals and semiconductors has resulted in a comprehensive theory for the transport properties in the surface region. This theory makes it possible to understand some of the complex behavior observed in semiconductors and metal due to interactions of the charge carrier with the surface layer. These effects become of increas-

ing importance as technology leads to greater use of thin film semiconductors or metal in microminiaturization of electronic circuits.

Research in metallurgy includes studies of the crystal structure of the family of alloys TiFe, TiCo and TiNi. A unique martensitic transition was found in all these metals. We also found that the intermetallic alloy Ti_2FeCo is superconducting at about 3.2 K. We are attempting to understand the unusual magnetic behavior of these metal alloys by means of this analysis.

We have now developed a general theory of magnetostriction, forced magnetostriction, and anomalous thermal expansion of ferromagnets of arbitrary symmetry. The theory has so far been successfully applied to Dy, Gd, and EuS.

An experimental study of anisotropy in the rare earth metals is being made in magnetic fields up to 140 koe. The results found so far on one of the rare earths, Tb, indicates a significantly larger anisotropy than is predicted theoretically from susceptibility data.

The antiferromagnetic structures of MnP, $EuTiO_3$ and Rb Mn Cl_3 have been established through neutron diffraction studies. Polarized neutrons are being used to increase the sensitivity of the measurements.

We are investigating the optical transmission of water in the vicinity of the green wavelength where water has its highest transmission. Ultimately we shall try to obtain experimental data on ocean water transmission down to very great depths.

High intensity laser radiation impinging on water was found to produce a shock wave suggesting interesting possibilities of air to underwater communication.

Electron irradiation treatments of low coercive force Ni-Fe alloys have resulted in extreme rectangularity in magnetic hysteresis loops. Such material properties are greatly desired in various nonlinear magnetic devices.

During the past year our publication rate continues to be high. A total of 35 articles were published in scientific and technical journals, 45 talks were given at technical society meetings, seminars or special meetings, and 8 patents were applied for or granted.

SEMICONDUCTORS

Scattering of Conduction Electrons By Localized Surface Charges

R. F. Greene

Several possible scattering mechanisms have been mentioned[1] in the literature but before this time none had been treated in any quantitative fashion. The scattering that arises from a random array of charged centers on a crystal surface has been analyzed. Using the Born approximation to the Schrodinger equation, the differential scattering amplitudes have been calculated. Using a formalism just derived[2], the energy and angle dependence of the Fuchs reflectivity p and kinetic specularity W_o were evaluated.

This problem differs from charged impurity scattering in the bulk because of two important surface effects, the "aspect effect" and the "termination effect". The latter is a partial suppression of the scattering matrix element which occurs for electrons with k-vectors at near grazing angles. For these electrons the termination of the wave function at the surface cuts down the overlap of the wave function with the scattering potential. The "aspect effect" is a simpler matter, consisting of the increase in the number of surface scatterers intercepted by an incident deBroglie wavefront of unit diameter as it is turned toward grazing incidence. This calculation, which is valid for semiconductors, semimetals, and metals with appropriate choice of physical parameters, gives a Fuchs reflectivity which is strongly angle dependent, and which is drastically different in magnitude from the kinetic specularity W_o. It also provides no scattering at all at glancing angles, and hence, as seen above, a surface mobility with no cusp.

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1 Greene, R. F., Surface Science 2, 101 (1964)

2 Greene, R. F., Phys. Rev. 140 (1965)

Angular Dependence Surface Scattering and Mobility

R. F. Greene
J. N. Zemel

Prior theories[1,2] of the surface mobility of semiconductors predicted the existence of a cusp in the surface mobility as a function of the surface potential excess (band-bending), provided that the surface is strongly scattering and that the mean free path is longer than the screening length. Recent experimental studies (by Davis[3] and Frankl-Carmean[4]) show, however, that the cusp is absent on some surfaces and present on others, so that something was lacking in the theory.

This problem was resolved[5] by introducing the possibility of an angular dependence in the surface scattering. It was shown that a surface mobility cusp should occur if and only if electrons incident at near grazing angles are strongly scattered. It was pointed out that an experimental study of the occurrence or nonoccurrence of surface mobility cusp on a given surface provides the first means of studying the details of surface scattering mechanisms.

Boundary Conditions for Electron Distribution

R. F. Greene

The physics of the surface and its scattering processes goes into transport theory largely through the boundary condition on the distribution function which Fuchs[6] introduced in 1938, and which has since then been universally used for metals, semimetals, and semiconductors. The single Fuchs reflectivity parameter p, which has to represent the scattering properties of the surface, has up to now been treated as an adjustable parameter. Unanswered all this time have been the deeper questions of the validity of the Fuchs b.c., and of how to tie the Fuchs reflectivity to scattering events.

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[1] Greene, R. F., Frankl, D. R., and Zemel, J. N., Phys. Rev. 118, 967 (1960)

[2] Greene, R. F., Phys. Rev. 131, 592 (1963)

[3] Davis, J. L., Surface Science 2, 33 (1964)

[4] Frankl-Carmean, Private Communication

[5] Greene, R. F., Phys. Rev. 140 (1965)

[6] K. Fuchs, Proc. Cam. Phys. Soc. 34, 100 (1938)

These questions have been answered[1] by setting up a fundamental theory of the boundary condition for electron distributions at crystal surfaces, based on the simplest basic properties of electron wave functions at surfaces, and of conservation of electron fluxes. A new boundary condition has been derived which reduces to the Fuchs form under definite circumstances, but which takes a different form when a magnetic field or strong band bending is present. A formula for the Fuchs reflectivity p is given in terms of scattering amplitudes. It is shown that p is physically and numerically distinct from the kinetic specularity W_o (= probability of no scattering at the surface). This provides the general formalism with which any scattering model may be used in transport calculations in semiconductors, semimetals, and metals.

Band Structure of Tin Telluride

R. S. Allgaier
J. R. Burke, Jr.
B. B. Houston, Jr.

SnTe is a cubically symmetric material in which large deviations from stoichiometry result in unusually high carrier concentrations (p) for an exgrinsic material. Presently attainable values of p lie in the range from about $5 \times 10^{19}\,cm^{-3}$ to $1.2 \times 10^{20}\,cm^{-3}$. At these concentrations, large deviations from spheroidal energy surfaces in momentum space are expected. In addition more than one band is likely to be populated. One of the best techniques for deriving the band structure and determining the shape of the Fermi surface, in such a complicated situation, is to study the periods of the oscillations in the resistivity (Shubnikov-deHass effect) that occur at high magnetic fields. These oscillations are periodic in the reciprocal of the magnetic field intensity, and the period of the oscillation is inversely proportional to an extremal cross-sectional area of the Fermi surface perpendicular to the magnetic field.

Because of the high magnetic fields needed for SnTe, measurements have been made at the Naval Research Laboratory. A rotating sample holder has been designed so that measurements as a function of the crystallographic orientation of the magnetic field can be made in a solenoid field. Oscillations have been observed in all orientations and over the whole carrier concentration range studied, i.e., $5 \times 10^{19}\ cm^{-3}$ to $5 \times 10^{20}\ cm^{-3}$. At $P = 7.8 \times 10^{19}\ cm^{-3}$ for example, the results show that in all orientations, several periods are simultaneously present. The valence band therefore cannot be described by the <111> ellipsoid of revolution model suggested by preliminary results. When samples with $p \geq 2.5 \times 10^{20}\ cm^{-3}$ were measured, a new, much longer period oscillation appeared. This period was not present

[1] Greene, R. F., Phys. Rev. 140 (1965)

at $p = 1.6 \times 10^{20}$ cm^{-3}. The Fermi level therefore enters a second valence band at a carrier concentration between 1.6 and 2.5×10^{20} cm^{-3}. Fig. 1 shows the new oscillation together with an oscillation resulting from the carriers in the first band.

Fundamental Absorption Edge of Tin Telluride[1]

E. G. Bylander
J. R. Dixon
H. R. Riedl
R. B. Schoolar

Recent studies of the electrical and thermal properties of p-type tin telluride, SnTe, indicate that the material is a semiconductor. The similarity of its electrical properties to those of the well-known semiconductor lead telluride, has been demonstrated and adds additional weight to the belief that the material is a semiconductor. However, the possibility that SnTe is a semimetal could not be definitely ruled out on the basis of prior information. The existence of a fundamental optical absorption edge is of considerable pertinence to this uncertainty in classification. This arises from the fact that such an edge is associated with a forbidden energy gap between conduction and valence bands. Such an energy gap is a necessary characteristic of all semiconductors.

Optical measurements in the infrared region were carried out in an effort to identify a fundamental absorption edge in SnTe. The major difficulty of such measurements in the past has been that extremely thin samples are required because of the large optical absorption existing throughout the infrared region. This absorption is at least partially due to the high concentration of free carriers which characterizes SnTe. This difficulty was overcome by preparing large-area, thin single-crystal films of SnTe grown on cleaved potassium chloride. Using these films, the sharp absorption edge shown in Fig. 1 has been observed in the infrared region near 0.5 ev. This edge is believed to be associated with the onset of direct transitions between a valence and a conduction band. On the high energy side of the minimum seen in Fig. 2, the absorption increases rapidly toward a plateau region indicated by the results of Cardona and Greenaway. This behavior is characteristic of a fundamental absorption edge. The increase in absorption on the low energy side of the minimum is believed to be associated primarily with free-carrier absorption. The results therefore lead us to believe that SnTe is a semiconductor.

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[1] E. G. Bylander, J. R. Dixon, H. R. Riedl and R. B. Schoolar, Phys. Rev. 138, A864 (1965)

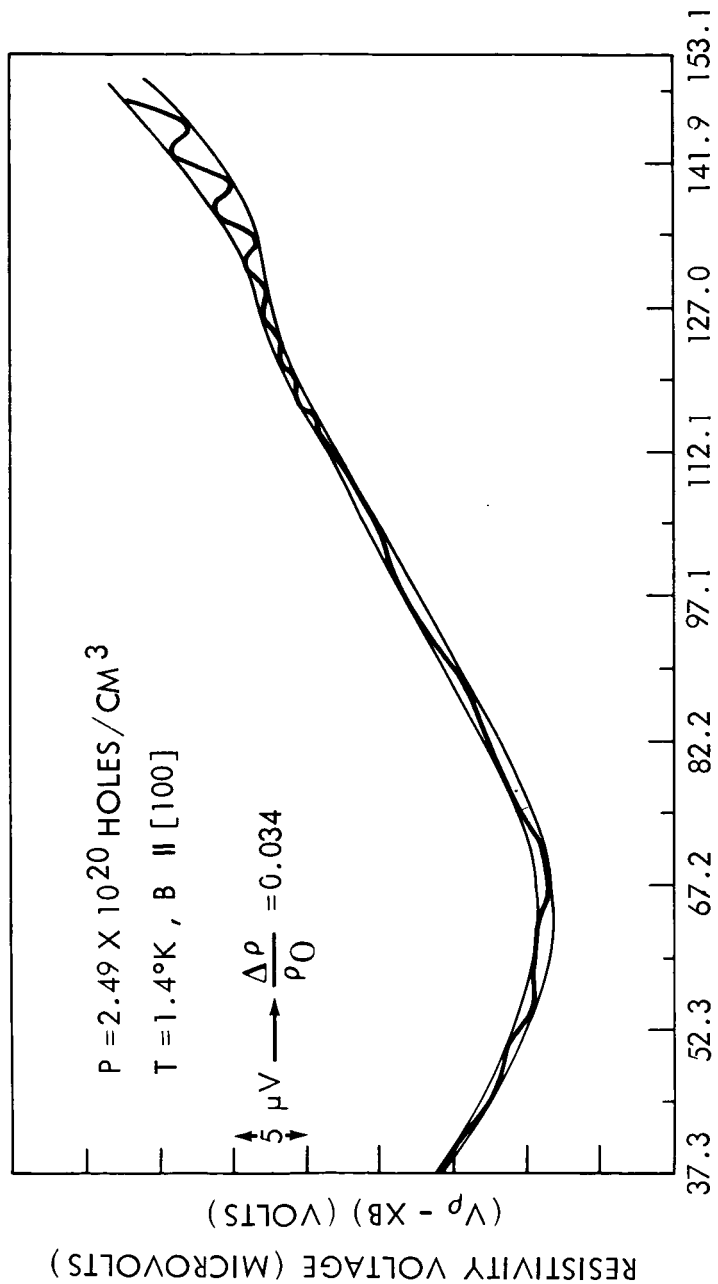


FIG. 1 MAGNETORESISTANCE IN P-SnTe

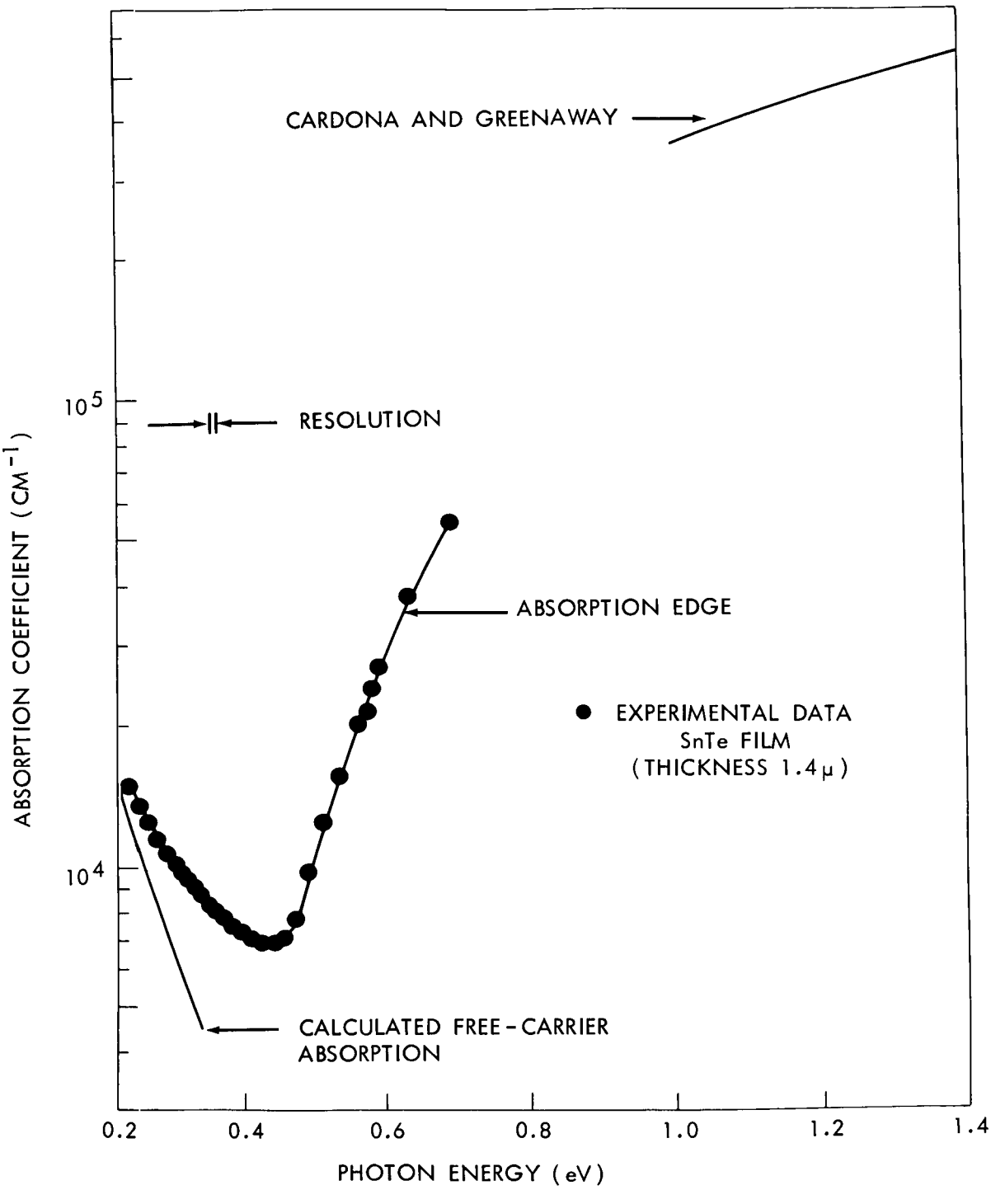


FIG. 2 OPTICAL ABSORPTION OF p-TYPE SnTe AT ROOM TEMPERATURE FOR MATERIAL HAVING A CARRIER CONCENTRATION OF 1×10^{20} cm^{-3}.

Point Defects in Tin Telluride

R. S. Allgaier
E. Gubner
B. B. Houston
M. K. Norr

SnTe crystallizes in the NaCl lattice. If this lattice is perfect, the crystal contains 50 atomic percent tellurium. However, SnTe crystals can be prepared with as much as 51 atomic percent tellurium. This implies that these crystals contain large numbers of point defects, which could be either vacancies, missing atoms from the Sn sites or intersticials in the lattice. To understand completely the properties of SnTe which are affected by the point defects the nature of the defects must be known. These properties include the electrical properties, mechanical properties and others of interest. The technique to determine the nature of the defects involves the use of precision lattice parameter, density, and composition determinations. From the lattice parameter the number of normal sites in the crystal can be computed. Since the weights of the atoms are known, the density the crystal would have if all the sites are occupied can be computed. If it is assumed that the deviation occurs by means of a particular type of defect, a computed correction to that density from the composition will tell how many atoms must be introduced or removed from the perfect crystal to give the proper composition. A comparision of this computed density with the measured density confirms or refutes the assumption about the type of defect involved. Since there are large numbers of defects in SnTe, the resulting discrepancy from choosing the wrong model is relatively large.

Point defect measurements are restricted to small samples because the sample composition is varied by diffusion. Techniques have been developed for precision density determinations on these small samples and a number of high quality single crystal samples covering a wide range of compositions have been prepared. Methods for precision lattice parameter determinations on these crystals are being developed since powder techniques are not reliable. The study confirmed the fact that the predominate point defects in SnTe are vacancies. We have yet to show that we can fit this model to very precise data over a wide range of compositions. Our data does show that the density does not change very much with composition while the lattice parameter changes considerably. This implies that there is considerable collapse of the lattice into the vacancy. This is contrary to theoretical predictions based on ionic bonding and symmetrical relaxation around the vacancy. This suggests that one should look for asymmetrical relaxation by means of internal friction studies in the kilohertz region

The Effect of Annealing on Dislocation Density in Lead Telluride

E. Gubner
B. B. Houston
M. K. Norr

In order to study the effect of various defects on the electrical properties in the lead chalcogenides we have been engaged in a program to produce single crystals of these materials as free from defects as possible. Highly perfect material is also extremely useful for studies of many of the intrinsic properties of these materials. Etches have been developed to determine the dislocation density in PbTe and methods have been developed for growing PbTe with the lowest dislocation densities which have been reported in this material. The effect of annealing on the dislocation densities in these crystals has also been studied in an effort to produce still lower dislocation densities.

The dislocation densities in these crystals can be lowered by a factor of from 5 to 10 by annealing at a temperature about 30° C below the melting point for 100 hours. PbTe of the composition used in these studies melts at 925° C. Typical results are shown in Fig. 3. The anneals are carried out in evacuated quartz tubes. The vapor pressure of PbTe at these temperatures is several Torr. The high vapor pressure makes these experiments difficult because the crystal can evaporate completely in much less than 100 hours if there is a cool spot on the tube where the vapor can condense. A major effort in this study has gone into building annealing furnaces with sufficiently uniform temperatures to prevent the evaporation of the crystal.

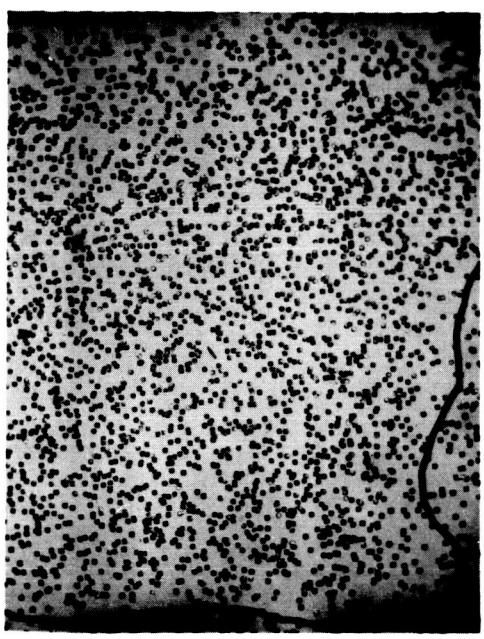
Cyclotron Resonance of Epitaxial Films

J. D. Jensen
J. N. Zemel

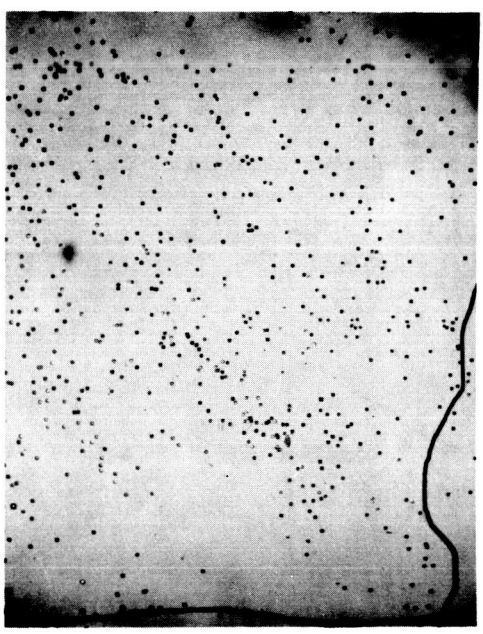
In the presence of a dc magnetic field, a free charge rotates about the direction of the magnetic field with an angular frequency

$$\omega_c = + \frac{eH}{m^* c}$$

where ω_c is the cyclotron frequence and e is the charge and m* is the effective mass of the particle. If a microwave field is now applied perpendicular to the magnetic field resonant absorption may occur when the microwave angular frequency equals ω_c. The physical requirement for observing this resonance is $\omega\tau \geq 1$, where ω is the microwave angular frequency and τ is the average time between collisons. This condition is satisfied in high quality epitaxial films of the lead salts at liquid helium temperature.



(a) BEFORE ANNEALING



(b) AFTER ANNEALING

FIG. 3 CRYSTAL WITH INITIAL DISLOCATION DENSITY OF 2×10^5 cm^{-2}. BEFORE AND AFTER ANNEALING.

PHOTOGRAPHS OF PbTe IN WHICH THE PITS OCCUR AT POINTS WHERE DISLOCATIONS INTERSECT THE SURFACE. (a) SHOWS A CRYSTAL WITH A DISLOCATION DENSITY OF 2×10^5 cm^{-2}, BEFORE ANNEALING, (b) SHOWS THE SAME AREA ON THAT CRYSTAL AFTER ANNEALING. THE DISLOCATION DENSITY HAS BEEN REDUCED TO 4×10^4 cm^{-2}. THE MAGNIFICATION IN THESE PHOTOGRAPHS IS ABOUT 50X.

A resonance in n-type PbTe at 4.2°K has been observed using a TE_{111} cylindrical cavity with a resonant frequency of 70 kmcs. The derivative of the power absorption is shown in Fig. 4. The value of H to be inserted into the equation is determined by averaging the maximum and minimum of the dP/dH curve. The values 0.061 and 0.083 for the effective mass agree very well with the results of Nii[1] on bulk n-type PbTe for the so-called dielectric anomaly. The low value of $\omega\tau$ ($\simeq 2$) for this particular film caused the true cyclotron resonance to be masked out. Also the thickness of the film was large compared to the cyclotron radius precluding the observation of any size effects.

Lead Salt Alloy Films

J. R. Dixon
E. Wenzel
J. N. Zemel

In the past four years, thin single crystal films of the lead salts, PbS, PbSe and PbTe have been studied. It has been shown that the properties of these films compare favorably with those of bulk materials. A natural extension of this work is to consider more complicated systems such as alloys of the above materials.

Single crystal-single phase thin film alloys of $PbTe_xSe_{1-x}$ have been produced over the entire composition range. These thin films were produced by co-evaporation of PbSe and PbTe in a system which has a separately pumped source and deposition chamber, thus eliminating contamination of the substrate during the initial warm up and pump down. The crystal perfection of a film of $PbTe_{0.5}Se_{0.5}$ produced by this method is shown in the Bragg diffractometer recording, Fig. 5. The Bragg angles for PbSe and PbTe are indicated and show that only one phase is present, the $PbSe_{0.5}Te_{0.5}$.

Germanium-Epitaxial Lead Sulfide Heterojunctions

J. L. Davis
M. K. Norr

This study began with a prior comparison of the photoconductive properties of the vacuum evaporated epitaxial PbS films, grown at NOL, with PbS polycrystalline films deposited chemically on glass. The polycrystalline films have poor crystalline perfection and the carrier's mobilities are two orders of magnitude lower than for single crystal material. It was hoped that the higher carrier mobility and high degree of crystalline perfection of the epitaxial films would yield superior photoconductive

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[1] Riro Nii, Rev. Elect. Comm. Lab., Tokyo 12 No. 11-12, Nov-Dec 1964

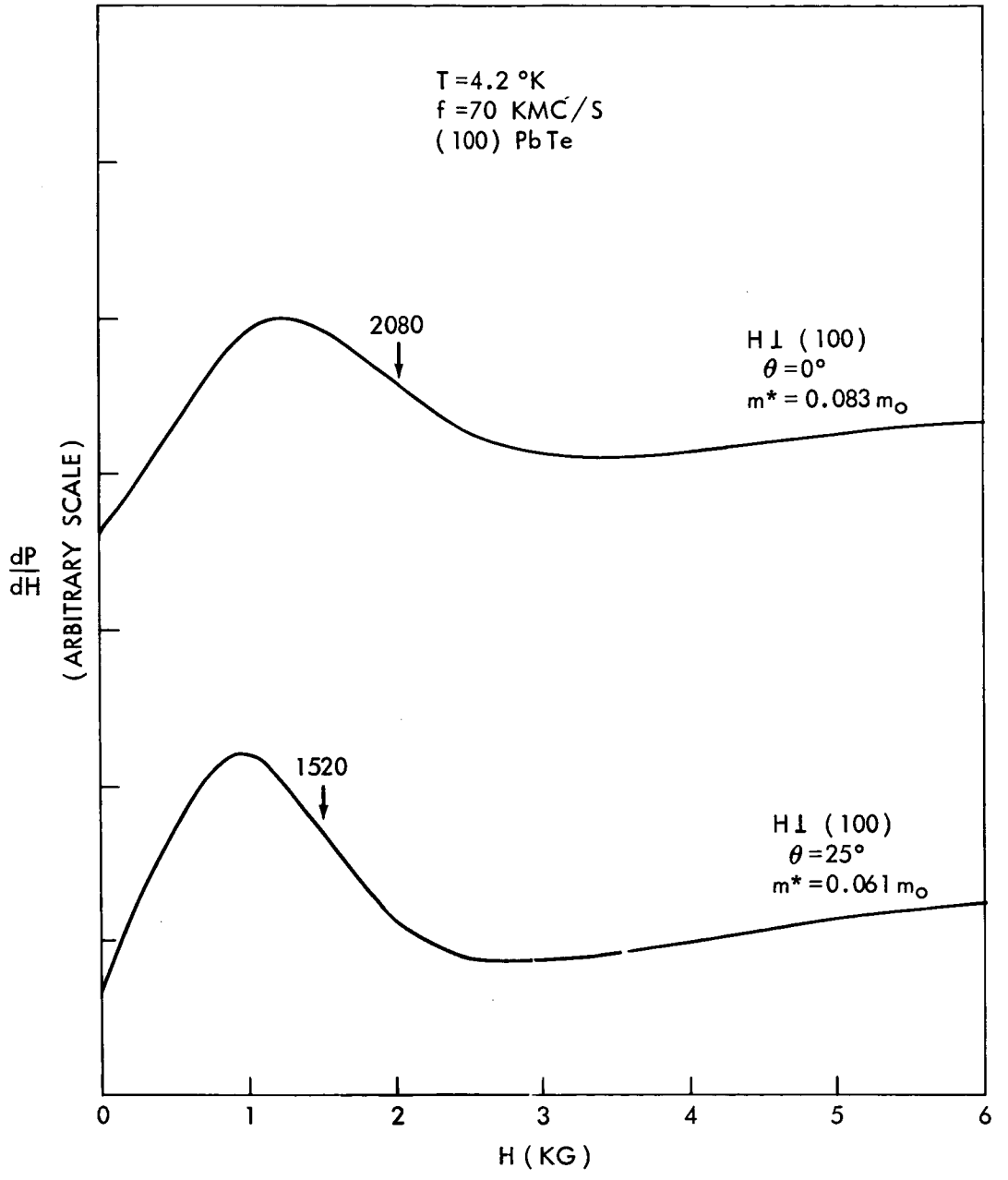

FIG. 4 THE DERIVATIVE OF THE CYCLOTRON RESONANCE POWER ABSORPTION VS MAGNETIC FIELD FOR n-TYPE PbTe

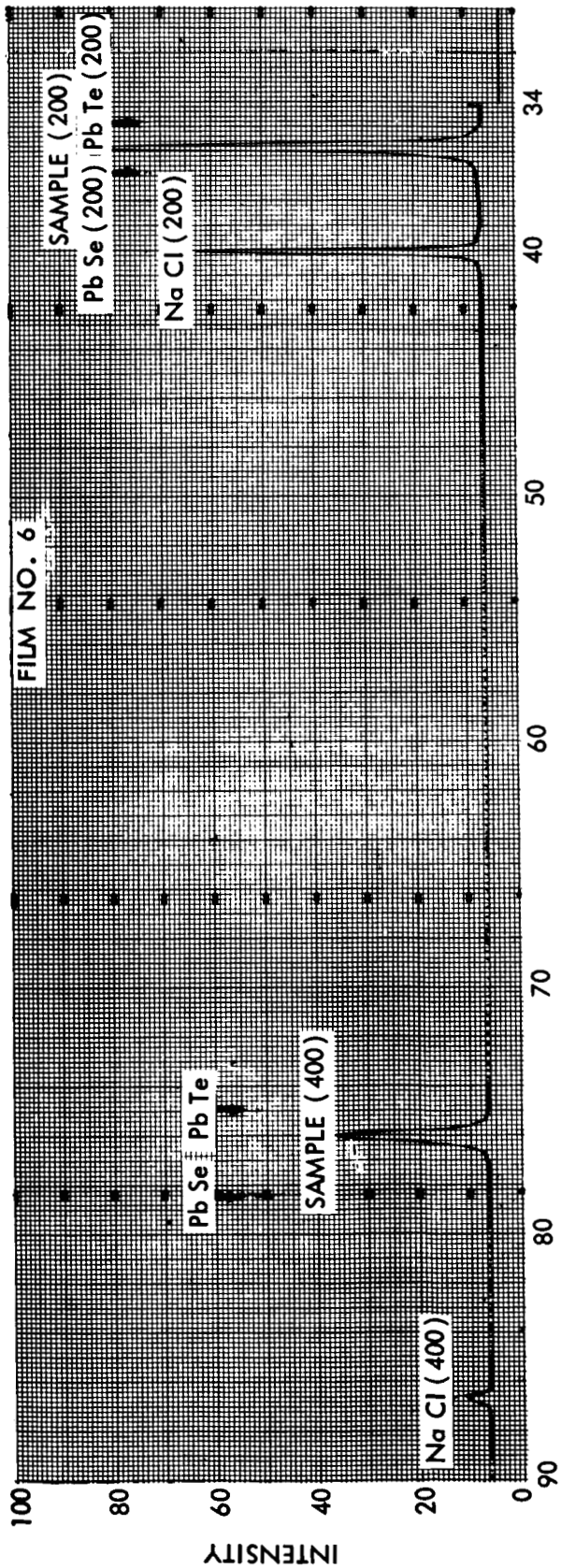

FIG. 5 BRAGG DIFFRACTOMETER PLOT OF EPITAXIAL FILM OF $PbSe_{0.5}Te_{0.5}$

detectors. However, this was not the case; the epitaxial films showed no photoconductive response. An attempt was then made to grow an epitaxial film from solution rather than by vacuum evaporation, in the hope of combining the advantages of both processes.

A Ge substrate and the lead nitrate-thiourea chemical deposition process was used. The substrates were cut, polished and etched and then had a PbS film deposited on the surface. The PbS film was checked for crystalline perfection by Laue back reflection and by the Bragg diffractometer. These results showed that the films were indeed epitaxial, i.e., they had the orientation of the substrate and they had the lattice spacing of bulk lead sulfide. The junction between the Ge and the PbS was tested as a photovoltaic device by illuminating it with monochromatic light through the Ge substrate. An appreciable response was found in the range from 1.5 to as far as 3.2 microns on some of the samples. The measured time constants for these junctions were shorter than 25 microseconds, for all cases. This is about two orders of magnitude faster than the PbS photoconductive detectors. Thus, these results indicate that epitaxial films can be grown from solution on semiconductor substrates and the junctions now obtained offer promise as fast infrared photovoltaic detectors.

MAGNETISM

Magnetostriction Theory

E. R. Callen

A general theory[1] of magnetostriction, forced magnetostriction, and anomalous thermal expansion of ferromagnets of arbitrary symmetry has been formulated using group theoretical arguments. It is based upon symmetry considerations imposed on the magnetoelastic Hamiltonian in contrast to the former classical approaches in which the symmetry is imposed at the level of the phenomenological free energy. Included in the Hamiltonian are both single-ion and two-ion interactions. The theory culminates in expressions relating the microscopic magnetostriction coefficients to the product of microscopic magnetoelastic coupling constants and certain spin correlation functions. Only three distinct types of correlation functions enter the theory; the self-correlation function $\langle (S_1^z)^2 \rangle$, the isotropic correlation function $\langle S_1 \cdot S_2 \rangle$, and the longitudinal correlation function $\langle S_1^z S_2^z \rangle$. From these functions the entire temperature dependence and the magnetic field dependence of the magnetostriction coefficients are derived.

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[1] Magnetostriction, Forced Magnetostriction and Anomalous Thermal Expansion in Ferromagnets, E. Callen and H.B. Callen, Phys. Rev. 139, A455, Jul 1965

Various approximation schemes are used to evaluate these spin correlation functions and to permit the application of the theory to specific materials. Successful applications of the theory have been made to Dy, to Gd and to EuS.

Dynamics of the Indirect-Exchange Model

E. R. Callen
A. Luther

Transport properties, such as electrical resistance, thermal conductivity and Hall effect of magnetic metals suggest a different type of magnetic structure than do the magnetic properties. Thus it is not known whether the magnetic moment is localized, as in an insulator, or whether it is distributed over the metal as a conduction electron. To learn more about magnetic metals, a simple model of a magnetic metal has been studied in detail. The model is the indirect interaction model, is similar to the type of interaction found in the rare earth metals. It assumes isolated spins at particular lattice sites, which are coupled to each other by the conduction electrons.

The two-time temperature-dependent Green's function method was used to study this model. The results can be given by an effective Hamiltonian of the form:

$$H = \sum_{i \neq j} \int_{-\infty}^{\circ} J_{ij}(\tilde{t})\, \vec{S}_i(t) \cdot \vec{S}_j(t-\tau)\, d\tau$$

This Hamiltonian is similar to the ordinary Heisenberg Hamiltonian, except for the appearance of retardation in the spin interactions. Physically, this means there is a delay in interaction between two spins because the conduction electrons which couple them can propagate the interaction only at a finite velocity. The propagacion characteristics are highly dispersible and can be quite slow at certain frequencies. The quantity $J_{ij}(\tau)$ depends on the details of the conduction electrons. We have calculated it for an interacting electron gas, and find the retardation corrections very important.

From the properties of the Green's function, formulae have been developed for the spin wave dispersion relation, temperature dependence of the magnetization, and many-body effects. Although analysis of these results is not yet complete, the temperature dependence of the magnetization caused by many-body effects has been ascertained. These effects are large in some magnetic metals, and raise questions about the spin-wave picture of magnetism.

Magnetostriction in the Heavy Rare Earth Garnets

R. M. Bozorth
A. E. Clark
B. F. DeSavage

The single-ion theory of magnetoelastic coupling by Callen and Callen has successfully accounted for the temperature dependence of the magnetostriction of some of the ferromagnetic rare earth metals[1] and the ferromagnet insulator yttrium iron garnet.[2] According to the theory only one magnetoelastic coupling constant per magnetic sublattice (the 0°K magnetostriction) is required to specify the entire temperature and field dependences of the magnetostriction. It is essential to know these coupling parameters in order to construct an atomic theory of magnetoelastic coupling from first principles.

The two magnetostriction coefficients, $\lambda^{\gamma,2}$ and $\lambda^{\epsilon,2}$ of single crystals of dysprosium, holmium and erbium iron garnets were measured using strain gage techniques from liquid nitrogen temperature to room temperature in fields up to 140 koe using the high field facilities at the National Magnet Laboratory, Cambridge, Mass. A striking feature of these data is the sharp dip in the magnetostriction at the compensation point, which follows from the failure of the sublattice moments to remain parallel to the applied magnetic field precisely at this temperature. Another predominent feature is the very large forced magnetostriction which abruptly reverses sign at the compensation point. Extrapolated values for the 0°K magnetostriction according to the single-ion theory of magnetostriction along with the giant magnetostrictions of Dy, Ho and Er metals are given in Table 1. The magnetostriction of the rare earth garnets is very large[3], 50 to 1000 times that of YIG and only an order of magnitude smaller than that observed in the rare earth elements themselves, revealing a very large coupling of the rare earth ion to the strains in both the metals and garnets.

The 0°K magnetostriction of the heavy rare earth garnets has also been calculated from the strain dependence of the electrostatic interaction between the crystalline field and the anisotropic charge distribution of the rare earth ions. The results provide the correct order of magnitude for the magnetostriction constants as well as the change in sign between Ho and Er iron garnets observed for $\lambda^{\gamma,2}$.

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[1] A. Clark, B. DeSavage, R. Bozorth, Phys. Rev. 138, A216 (1965)

[2] E. Callen, A. Clark, B. DeSavage, W. Coleman, and H. Callen, Phys. Rev. 130, 1735 (1963)

[3] A. Clark, B. DeSavage, N. Tsuya, S. Kawakami, Eleventh Annual Conference on Magnetism and Magnetic Materials, Nov. 1965

Table I

0°K Magnetostriction Coefficients

	$\lambda^{\gamma,2}(0) \times 10^6$	$\lambda^{\epsilon,2}(0) \times 10^6$
YIG	- 1	- 8
DyIG	- 2100	- 830
HoIG	- 1400	- 330
ErIG	630	- 450
Dy	8,500	9,200
Ho	2,400	-
Er	-3,700	-

Conduction Electron Polarization — H. S. Belson

The theoretical understanding of the magnetic transition metals iron, cobalt, and nickel is incomplete. The role of the valence and magnetic electrons is especially open to question. Simple theories indicate that the conduction electrons (those electrons at the Fermi level) are highly polarized, while more complex theories indicate somewhat less polarization. An experiment has been performed to measure this polarization using a capacitor, one plate of which is a magnetic metal, and the other an ordinary metal. As one applies a magnetic field, voltages which appear can be ascribed to a shift in the Fermi level of the magnetic material. This shift will be proportional to the spin polarization at the Fermi level.

No such shift has been found, within the accuracy of the experiment contradicting a single earlier report of complete polarization in iron. The studies show that spurious voltages can occur from several sources, most notably from a magnetostrictive-piezoelectric interaction, especially if the capacitor in question is made of an extremely thin coating of iron. No shift was found in nickel or cobalt.

High Field Susceptibilities of Iron and Nickel

R. M. Bozorth
A. E. Clark
B. F. DeSavage

Previous attempts to measure the high field magnetization of iron and nickel showed no certain increase in magnetization above 10 to 20 koe, the uncertainty corresponding to a volume susceptibility κ of $\pm$ 2 to 8 x 10^{-5}. In view of the relation of the high field susceptibility to the band structure in Fe and Ni, and attempt has been made to increase the accuracy previously attained.

The volume susceptibility in fields of 15 to 120 koe was measured by several techniques using magnet facilities of the IBM Watson Research Laboratories, the Naval Research Laboratory and the MIT National Magnet Laboratory. Results for Ni (at 78°K and 4°K) gives $\kappa = 5.5 \pm 1 \times 10^{-5}$ and for Fe (at 78°K only) $\kappa = 14 \times 10^{-5}$. Theoretical estimates of the Holstein-Primakoff susceptibility and the orbital paramagnetism indicates for Ni that the Pauli part of the susceptibility is $\cong 4 \times 10^{-5}$. This is about ten times smaller than what one would expect on the assumption of a full up-spin d band which implies a smaller exchange splitting and/or a wider d band than most current estimates. In Fe, there are certainly d-like holes in both spin-up and spin-down bands.

Anisotropy of the Rare Earth Metals

A. E. Clark
J. J. Rhyne

An experimental study of the magnetocrystalline anisotropy has been started. In the investigation of the basic magnetic and magnetoelastic properties of rare earths Dy, Dr, Ho, and Tb, the anisotropy energy has the following form:

$$E_k = K_2P_2(\cos\theta)+K_4P_4(\cos\theta)+K_6P_6(\cos\theta)+K_6^6P_6^6(\cos\theta)\cos 6\theta$$

where the P_1^m are Legendre polynomials and the K_1^m are the anisotropy constants. Angles θ and ϕ are, respectively, the directions of the magnetization with respect to the c and a axes. It has been shown that appropriate choices for the signs and magnitudes of the K's can account for the various observed spin structures and for the transitions between these phases. It is the purpose of the current study to determine experimentally the magnitude and temperature dependence of these anisotropy constants. It is hoped that the Callen one-ion theory, which was highly successful in predicting the temperature dependence of the basal plane magnetostriction in Tb and Dy, will also provide the correct explanation for the temperature dependence of the

anisotropy constants. Correlation of the experimental results with this theory would provide additional support for the apparent highly localized nature of the magnetic interaction in the rare earths.

The anisotropy is being determined from measurements of the torque on the specimen as a function of the angle of the applied magnetic field relative to the crystal axes. On the basis of a computer simulation of the experiment using best available estimates of the anisotropy, sufficient torque sensitivity has been achieved by mounting the sample on the end of a thin wall brass torsion tube whose shear is detected by strain gages. The anisotropy torque has been measured using a "dry-tail" dewar and automatic temperature control apparatus which allow temperature control from above room temperature to He temperature. The unusually high anisotropy precludes sufficient deflection of the magnetic moment in fields less than 100 koe. High field facilities up to 140 koe at the National Magnet Laboratory and the Naval Research Laboratory are being employed for the experiment. Initial data on Tb indicates an anisotropy of order 2×10^8 ergs/cm^3 at 150°K. This is significantly larger than that predicted theoretically on the basis of susceptibility data.

Magnetostriction in Single Crystal Terbium

A. E. Clark
B. F. DeSavage
J. Rhyne

The measured magnetostrictive strains in the single crystals of the heavy rare earth metals terbium, dysprosium, holmium, and erbium were found to be extraordinarily large 10^{-3} to 10^{-2} at low temperatures. When these strains are separated into those arising from spin operators, it is discovered that the lowest order shearing magnetostrictions arise primarily from a single-ion term in the interaction Hamiltonian and they have a temperature and field dependence given by:

$$\lambda(T,H) = \lambda(0)\, \hat{I}_{5/2} \left\{ \mathcal{L}^{-1}\left[M(T,H) \right] \right\}$$

Here $\lambda(0)$ is the 0°K magnetostriction, $\mathcal{L}^{-1}[M(T,H)]$ is the inverse Langevin function of the moment, and $\hat{I}_{\ell+\frac{1}{2}}$ is a reduced hyperbolic Bessel function of order $\ell+\frac{1}{2}$, where 2ℓ is the degree of the spin operators.

The four 0°K magnetostriction coefficients of Tb metal obtained from measurements in the paramagnetic region have been evaluated by applying the above relation. The enormous magnetic anisotropy below the ordering temperature precludes any measurement of these

essential parameters below~230°K. Tsuya, Clark and Bozorth,[1] predicted the 0°K values of these strains on the basis of the distortion of the crystalline field due to nearest neighbors and the electric field arising from the strained charge distribution of conduction electrons assuming a deformable ion model. Both of these atomic mechanisms give good agreement with the variation of the magnetostriction throughout the heavy rare earth series. By comparing the ratios of the four 0°K magnetostriction coefficients of Tb, it is possible to determine if any of these mechanism dominate. The calculated coefficients (normalized to 0.01 for $\lambda^{\gamma,2}$) are compared to the experimental measurements in Table II. The dominant source of the magnetostriction is obviously the interaction between the 4f electron charge distribution and that of the conduction electrons. The contribution to the magnetostriction of the crystalline field due to nearest neighbors, on the other hand, must be very small since it predicts a negative $\lambda^{\gamma,2}$ and a large $\lambda^{\alpha_2,2}/\lambda^{\gamma,2}$ ratio, which is not observed.

Table II

Comparison Between Experiment and Theory
Of Magnetostriction Coefficients of Terbium

λ	Experiment	Deformable Ion	Crystal Field
$\lambda_1^{\alpha,2}$	-.0026	-.0045	-.027
$\lambda_2^{\alpha,2}$	.009	.009	.053
$\lambda^{\gamma,2}$	.010	.010	.010
$\lambda^{\epsilon,2}$	.0115	.009	-.037

Magnetic Structures by Neutron Diffraction — H. A. Alperin, S. J. Pickart

The cross section for elastic scattering of neutrons from a magnetic solid contains a term arising from the interaction of the neutron magnetic moment with the spin of the unpaired

[1] N. Tsuya, A. Clark, R. Bozorth, Proc. International Conf. on Magnetism, Nottingham, England, 1964

atomic electrons. Observation of this magnetic scattering, since it depends in a known way on the magnitude, periodicity, and orientation of the atomic spins, leads directly to determination of the magnetic spin arrangement in the material.

Powder neutron diffraction studies have been used to establish antiferromagnetic structures in $EuTiO_3$ (where initial magnetization studies suggested ferromagnetism), and in $RbMnCl_3$ (where antiferromagnetism had been inferred from susceptibility measurements). Single crystal measurements were performed on MnP, in which low temperature magnetization measurements had indicated metamagnetism, i.e. an antiferromagnetic-to-ferromagnetic phase change upon application of an external field. The neutron diffraction measurements show that the zero-field structure is in fact a spiral of rather general type, in which the spins related by a center of symmetry remain parallel and the spiral is propagated by a rotation of 20° for every operation of the twofold screw axis parallel to the orthorhombic c axis.

Spin Wave Dispersion in Ferromagnetic Metals

H. A. Alperin
S. J. Pickart

Measurements described in last year's report of spin wave dispersion in ferromagnets by the inelastic scattering of polarized neutrons have been continued. It may be recalled that in these measurements one is seeking to determine the basic nature of the exchange forces acting in ferromagnetic metals by examining the changes in energy and momentum of scattered neutrons when spin waves or magnons are excited. Polarized neutrons are used to increase the sensitivity by eliminating unwanted contributions from phonon scattering; however, the available polarized neutron flux did not allow energy analysis of the scattered beam, so that the dispersion curve could not be measured directly. For a given misset of the crystal from the Bragg position (e.g. a given energy change) a sharp cutoff corresponding to a particular magnon wave vector is observed as the scattering angle is varied (see Fig. 6). The observations as a function of misset angle then trace out scattering surfaces whose shape may be least-squares fitted by varying the parameters of a theoretically expected dispersion curve.

The results[1] of this procedure for body-centered cubic Fe are shown in Fig. 7. It will be noted that the dispersion curve derived from our data departs considerably from a quadratic form $\hbar\omega = Dq^2$ (where $\hbar\omega$ is the spin wave energy and q the wave vector) such as had been previously predicted. The results can however be fitted by including higher order terms in q^2; the

[1] G. Shirane, R. Nathans, O. Steinsvoll, H. Alperin, S. Pickart, Phys. Rev. Ltrs. 15, 146 (1965).

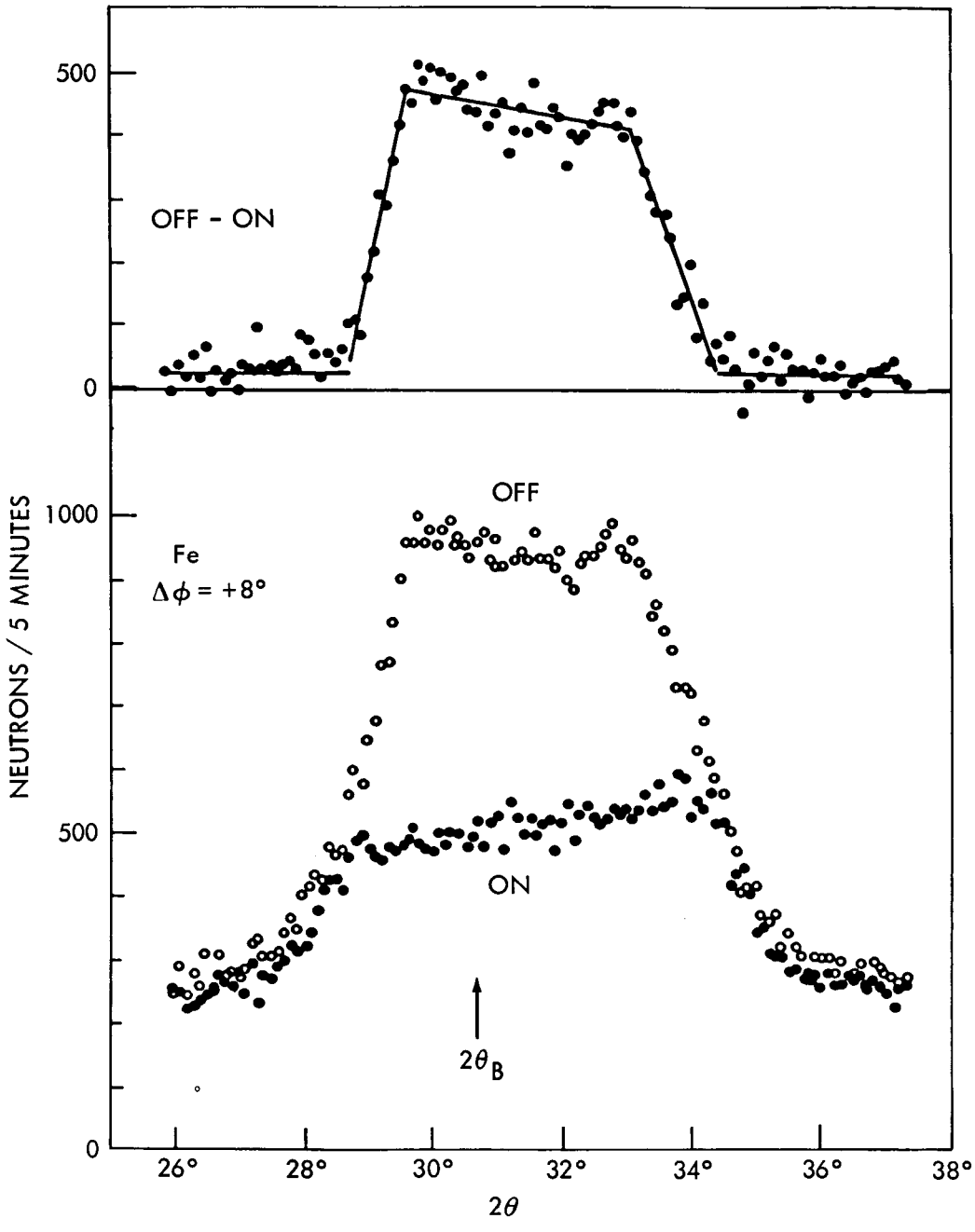

FIG.6 THE SHARP CUTOFFS CAUSED BY A MAGNON SCATTERING SURFACE AS THE NEUTRON DETECTOR IS SCANNED THROUGH A RANGE OF SCATTERING ANGLES 2θ FOR A GIVEN ANGLE OF CRYSTAL MISSET, $\Delta\phi$. ($2\theta_B$ IS THE BRAGG ANGLE.) THE DIFFERENCE OF THE TWO SPIN STATES, LABELLED HERE AS THE R.F. COIL NEUTRON FLIPPER ON OR OFF, CONTAINS MAGNON CONTRIBUTIONS ONLY, WHILE THE ON STATE CONTAINS PHONON CONTAMINATION.

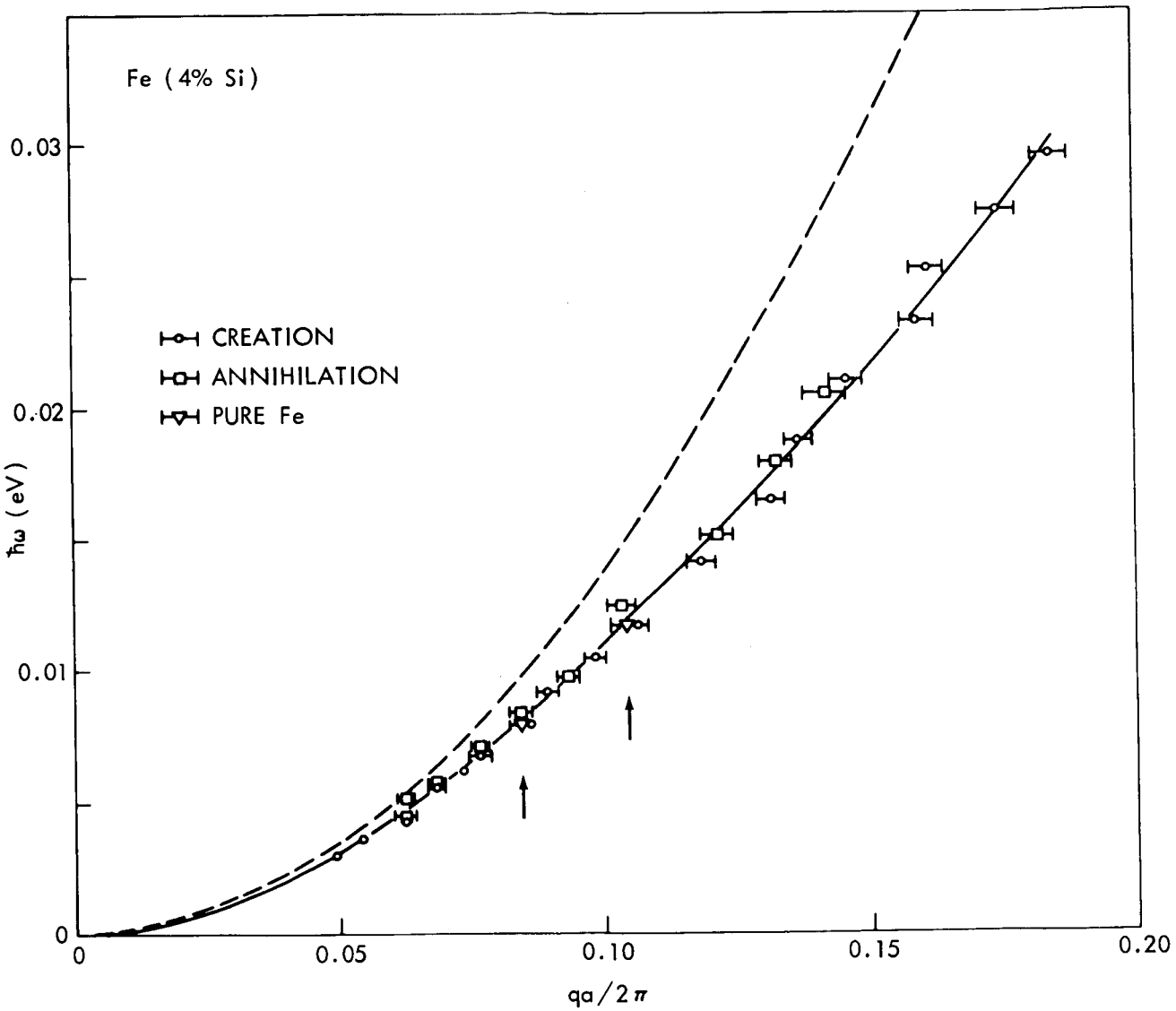

FIG. 7 MAGNON DISPERSION RELATION FOR IRON CONTAINING 4% Si. THE ARROWS CALL ATTENTION TO TWO POINTS MEASURED FOR PURE IRON. THE DASHED CURVE IS THE FUNCTION $\hbar\omega = 286\ q^2$ WHILE THE SMOOTH CURVE IS A LEAST-SQUARES FIT THROUGH THE EXPERIMENTAL POINTS AS EXPLAINED IN THE TEXT.

solid curve is an expression of the form $\hbar\omega = Dq^2(1 - \beta q^2)$ with $D = 266$ meVÅ^2 and $\beta = 3.2$Å^2. Similar results have been obtained for hexagonal close-packed Co, for which we find $D = 490$ meVÅ^2 and $\beta = 3.3$Å^2. Neither the Heisenberg nearest-neighbor model or the present band structure calculations predict as large a Q^4 term as observed here. The Heisenberg model can be reconciled with the data by assuming an arbitrary long-range interaction (presumably through conduction electrons) of about ten near-neighbor distances; or the band model by alteration of the density of states curve. Which of these alternatives is correct will become apparent as more detail in the form of the dispersion curves is discovered, particularly at higher q values than obtainable here, which correspond to three-tenths the distance to the Brillouin zone boundary.

Spin Wave Dispersion in Ionic Compounds

H. A. Alperin
S. J. Pickart

The spin wave spectra in ionic compounds should be more straightforward than in metals since they behave in general as though their moments are well-localized, so that a Heisenberg model should be applicable. Interest in determination of the exchange interactions then centers on comparison with predictions for these quantities made by various ordering theories on the basis of macroscopic quantities such as susceptibility, specific heat, transition temperatures, etc., and, more basically, with calculations of these interactions from first principles, starting from atomic wave functions.

Measurements have been obtained on two ionic compounds, ferrimagnetic Fe_3O_4 and antiferromagnetic $KMnF_3$. The first, Fe_3O_4, was carried out by the polarized neutron method described above, primarily as a means of testing the method; since a dispersion curve from inelastic scattering of unpolarized neutrons using energy analysis already exists. Very good agreement was obtained with this previous measurement by our indirect method. Antiferromagnetic $KMnF_3$ was measured on the Harwell twin-rotor time-of-flight spectrometer; a directly measured dispersion curve was obtained for q values reaching to the zone boundary, which could be fitted by nearest and next-nearest neighbor exchange interactions of 0.327 meV and 0.009 meV respectively. An energy gap of 0.8 meV at $q = 0$ was also inferred from the data, which is much larger than expected from measurement of the nuclear magnetic and electronic spin resonance on this material.

INTERMETALLICS

W. J. Buehler
H. E. Eberly
J. Goff
D. Goldstein
A. Rozner
C. Sutton
A. Syeles
F. Wang
R. Wiley

The crystal structures of TiFe, TiCo and TiNi have been determined through single crystal X-ray and powder neutron diffraction methods. A unique "martensitic" transition has been found to exist in these alloys: TiFe - below 4°K, TiCo - at 38°K and TiNi - at 439°K (Fig. 8). The unusual mechanical properties of TiNi, previously reported, are explained based on its 9Å superstructure (substructure has a 3Å repeat) and the "martensitic" transition. On the other hand, the drastic change in mechanical properties, brittle → moderately ductile → extremely ductile, in going from TiFe → TiCo → TiNi can best be explained on the basis of the cohesive energy contributed by free electrons (Fig. 9). This explanation points to the interesting possibility of raising the transition temperature, and hence the fracture strength, through addition of hydrogen as an interstitial electron donor. This possibility has been confirmed experimentally.

It was also determined that the intermetallic alloy Ti_2FeCo is superconducting with a transition temperature of approximately 3.2°K. It has been reported by Matthias et al of Bell Labs that TiCo was superconducting below 0.71°K but that TiFe was not, at least down to 0.30°K. The interesting point is that Prof. Beck of the University of Illinois has found that the alloys of these transition metals appear to have a band structure that depends upon their electron number (the number of electrons outside the last closed shell) per atom rather than the constituents. Thus, it would be interesting to investigate the superconducting properties of alloys of these transition metals with the same electron number.

Young and shear modulus as a function of temperature between -710° C and 400° C were determined by a dynamic method for polycrystalline TiNi intermetallic compound. Young's modulus exhibits a minima at 70° C. The modulus rises steeply with both increasing and decreasing temperature. The discontinuity of the modulus indicates occurrence of a first order transition in TiNi around 70° C.

The effect of cold working on mechanical properties of TiNi intermetallic compound (50 at .% Ni, 50 a .% Ti) was investigated. Samples were cold worked at temperatures well below their martensitic transformation temperature by means of conventional metal working techniques. The low temperature de-

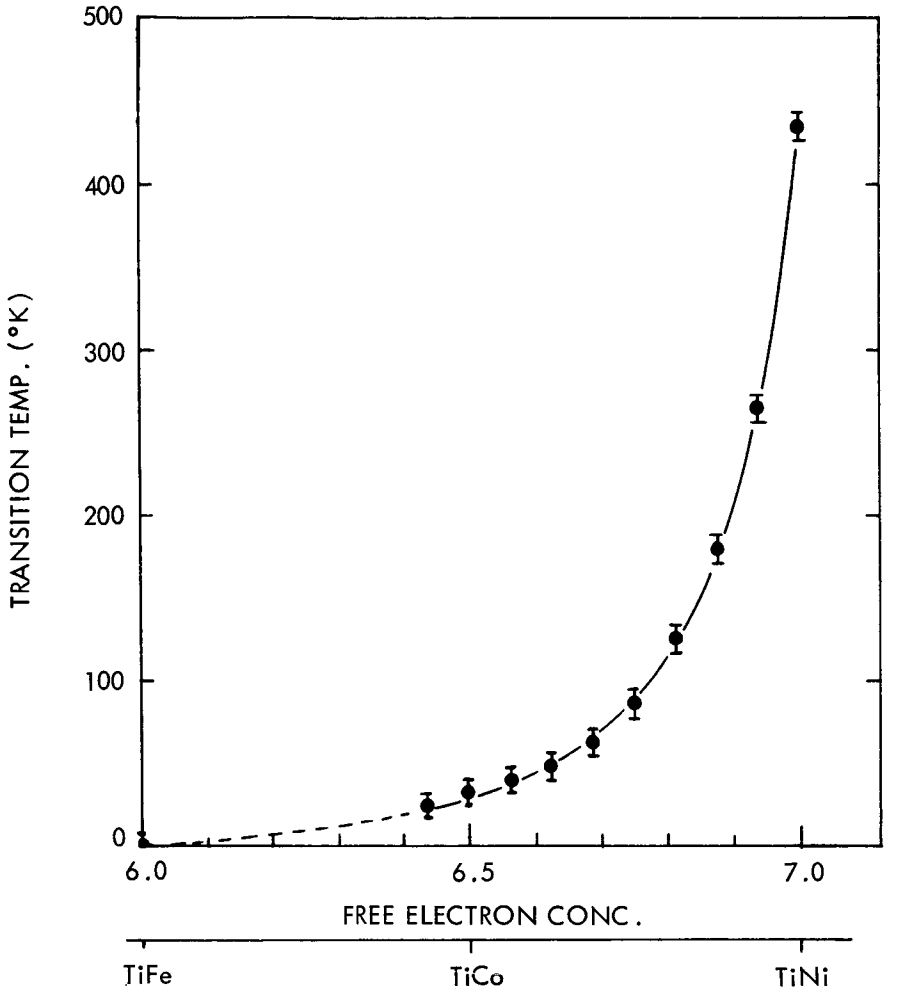

FIG 8 THE "MARTENSITIC" TRANSITION TEMPERATURES VS COMPOSITIONS IN TERNARY PHASES, Ti (Ni_x,Co_{1-x}), Ti (Co_x, Fe_{1-x}) THEREFORE, VS FREE ELECTRON CONCENTRATION

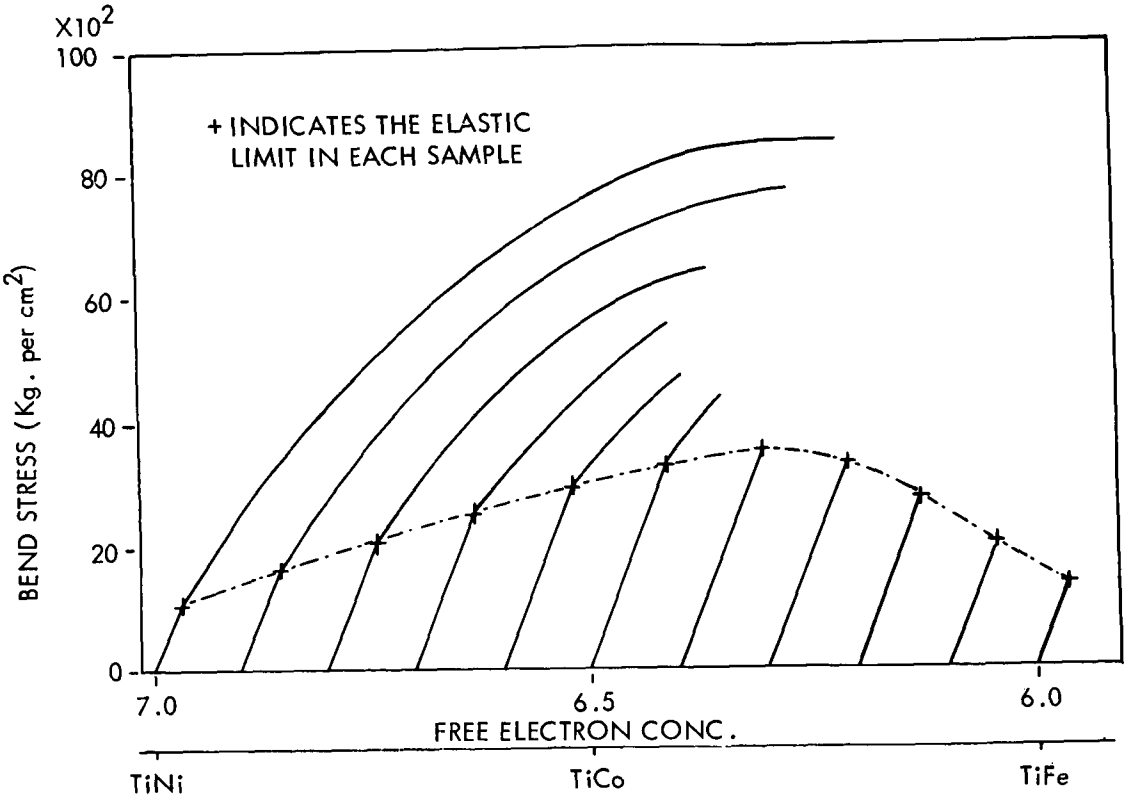

FIG. 9 BEND STRESS-STRAIN CURVES OF THE Ti-Ni-Co, Ti-Co-Fe TERNARY ALLOYS WITH THE "CsCl"-TYPE STRUCTURE. THE MEASUREMENTS WERE MADE ON A BALDWIN UNIVERSAL TESTER, USING 0.64 CM/MIN LOADING RATE AND AT ROOM TEMPERATURE. ALL SAMPLES WERE IN THE ARC-CAST CONDITION AND GIVEN A STRESS-RELIEF-ANNEAL AT 800°C FOR ONE HOUR PRIOR TO TESTING.

formation increases the yield strength from 18,000 to 61,000 psi and the ultimate tensile strength 125,000 to 220,000 psi while preserving elongation of 15 percent. The low temperature ductility is ascribed to a martensitic transformation and enables deforming the TiNi compound at low temperatures where the work hardening rate is very high. The martensitic transformation is promoted by the application of external forces and the TiNi martensite exhibits high strength and ductility at room temperature. This increased strength indicates the possible usefulness of the TiNi alloys for underwater structures.

An attempt was made to improve the elevated temperature strengths in the compound TiNi by ternary additions. Ternary alloys of the intermetallic compound TiNi were hot hardness tested. Although hardness can be increased by such alloying, neither the base material nor the alloys offer promise for long time service use above 800° F. Silicon, aluminum, iron, and cobalt are effective room temperature hardeners. A unique interfacial compatibility between TiNi and tungsten bulk metal was discovered. It is believed that this will enable introduction of a new metal-metal composite system of fibrous tungsten in a Nitinol matrix. A rod of such material is shown to be capable of room temperature swaging, suggesting improved methods of working highly refractory metals. Silver-free TiNi brazing alloys for joining tungsten metals are a natural result of such compatibility.

The Transport Properties of Transition Metal Alloys

J. Goff

When the number of electrons per atom in a metal is greater than one, its Fermi surface occupies more than one Brillouin zone and so is complex. It has been found by Beck and coworkers[1] that the alloys of the transition elements of the first long period with electron per atom ratio between that of Cr(6) and Mn(7) show the same trend in the coefficient of the electronic heat capacity irrespective of the constituents: Cr-Fe (ferromagnetic), Cr-Mn (antiferromagnetic), and TiX (non-magnetic). In particular there is a large peak at the ratio 6.4.

The thermoelectric power, electrical resistivity, and thermal conductivity of the TiX compounds and alpha-CrFe have been measured from about 2° to 300° K to determine their sensitivity to the complex Fermi surface and to search for similarities between the magnetic and non-magnetic systems.

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[1] K. P. Gupta, C. H. Cheng, and P. A. Beck; Metallic Solid Solutions, Benjamin, New York, 1963, p. XXV-1.

It appears that the Fermi surface of TiFe(6) is multiband while that of TiNi(7) is much simpler as would be expected from the simple nearly free electron filling of the Brillouin zones. The thermopower of TiFe(6) shows the same type of anomaly as is seen at somewhat higher temperatures in Cr(6). Thermal conductivity measurements on this compound and its alloys indicate that the anomaly involves phonon assisted hole transfer between bands in the Brillouin zone. In general the comparison of the transport properties of the non-magnetic and magnetic alloys appears to be fruitful. For example, the magnetic alloys show extremely anomalous resistivities for compositions which correspond to non-magnetic compounds that in turn have anomalous resistivities that can be analyzed as multiband departures from Matthiessen's Rule.

Superconductivity in the Titanium Alloys

J. Goff

It has been observed by Fowler[1] et al that all the elements in the center of the periodic table are either ferromagnetic, antiferromagnetic, or superconducting. That is, at some low temperature the electrons of these solids show some sort of cooperative phenomena. Hulm[2,3] and co-workers have investigated transition metal alloys and found that the nonmagnetic ones show a high incidence of superconductivity in the temperature range above 0.3°K. Matthias[4] has found that TiCo is superconducting with a transition temperature of 0.7°K.

In this Laboratory, we found that Ti_2FeCo is superconducting with a transition temperature of about 3.3 to 3.5°K and a transition temperature region of about 0.2°K. The existence of superconductivity in this compound tends to confirm the observations that these compounds are non-magnetic in spite of the presence of large amounts of ferromagnetic elements (Fe, Co, Ni). Based on the great increase in transition temperature above that found for TiCo, the next step is to seek the maximum and to try to understand it in terms of the band structure of these compounds that is being deduced from the other transport measurements

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[1] R. D. Fowler, B. T. Matthias, L. B. Asprey, H. H. Hill, J. D. G. Lindsay, C. E. Olsen, and R. W. White, Phys. Rev. Ltrs. 15, 860 (1965).

[2] J. K. Hulm and R. D. Blaugher, Phys. Rev. 123, 1569 (1961).

[3] S. H. Autler, J. K. Hulm, and R. S. Kemper, Phys. Rev. 140, A1117 (1965).

[4] B. T. Matthias, T. H. Geballe, and V. B. Compton, Rev. Modern Phys. 35, 1-22 (1965).

PROPERTIES OF GLASS

Temperature Dependence of the Velocity of Sound in SiO_2, GeO_2, B_2O_3, As_2O_3, As_2S_3, and As_2Se_3 Glass

R. P. Ryan
R. E. Strakna

Measurements of the temperature dependence of the longitudinal ultrasonic velocity were taken in SiO_2, GeO_2, B_2O_3, As_2O_3, As_2S_3 and As_2Se_3 glass from 1.5 to 400°K. In SiO_2, GeO_2, and B_2O_3, the rate of change of velocity with temperature increases with decreasing temperature in the liquid helium temperature region instead of approaching zero. In constrast, the velocity temperature dependence in As_2S_3 and As_2Se_3 resembles that of a crystalline solid. As_2O_3 behaves normally down to 77°K, the lowest temperature at which the change in velocity was measured in this glass. While SiO_2 and GeO_2 have an anomalous positive temperature dependence of the velocity at high temperatures, the other four glasses have a normal negative dependence.

This elastic study and the earlier analytical study of these glasses suggest that the diverse behavior of the temperature dependence of the ultrasonic velocity in simple glasses of the type AB_2 or A_2B_3 is due to the difference in the characteristics of the large relaxation loss in these glasses. That is, the presence of an extremely anharmonic optical mode of vibration associated with a barrier potential makes available vibrational energy states separated from the lowest states by energies between 1.0 and 100°K in terms of E/k for SiO_2, GeO_2, and B_2O_3, but not for As_2O_3, As_2S_3, and As_2Se_3. A change in the contribution to the elastic energy from this mode of vibration occurs at those temperatures where the upper of the close lying levels is being populated or depopulated with change in temperature. The calculated splittings of the energy levels also suggest a mechanism for a mechanical or dielectric resonance loss when the ultrasonic or r.f. measurement frequency is the same as the E/k separation of the levels.

APPLIED OPTICS

Light Transmission in Water

D. E. Matlack
W. W. Talbert
H. A. Templin

The availability of the laser high intensity light source and a simultaneous increase of naval underseas optical visibility and communication requirements has renewed our interest in the

nature of the transparency of water. One part of our comprehensive program to determine the optical characteristics of the ocean is concerned with the scattering coefficient resulting from inclusions in water. A second part is the determination of the true absorption of pure water, a parameter which sets an upper limit of expected performance for any system.

Our laboratory studies of total attenuation and scattering coefficients have enabled us to compute and report the first value of the absorption coefficient of highly filtered water, which is .056 m^{-1} at 4358Å. A series of measurements of the scattering coefficient were made for several types of water at selected wavelengths; and a spectral measurement of the attenuation coefficient was made in the visible spectrum for highly filtered water. The difference between the scatter coefficient, Fig. 10 and the attenuation was used to compute the absorption.

Acoustic Wave Generation by Lasers

C. E. Bell
B. V. Kessler
W. W. Talbert

High intensity laser radiation ($\sim 10^6$) watts when focussed into water, is capable of generating shock waves. A study has been initiated to generate shock waves in water using lasers and to determine the characteristics of these waves. The time development of an expanding shock wave, the thickness of the shock, density distribution behind the shock, temperature of the medium and other pertinent data are of interest. The existence of a shock and its velocity can be measured using an appropriate optical technique such as a Schlieren system.

Figure 11 shows a double pass-single mirror Schlieren system which was used to determine in a qualitative way, the magnitudes of thermal effects which take place long after the incident shock producing laser and the shock itself have been dissipated. The accompanying photo, taken with a 16 mm, 64 frames/sec camera, shows that a gradient in the refractive index in a direction perpendicular to the incident laser beam exists and has well defined boundaries. The thermal effect is still visible after 10 seconds. The effect in this case was produced with a long pulse (600 μ sec) neodymium laser.

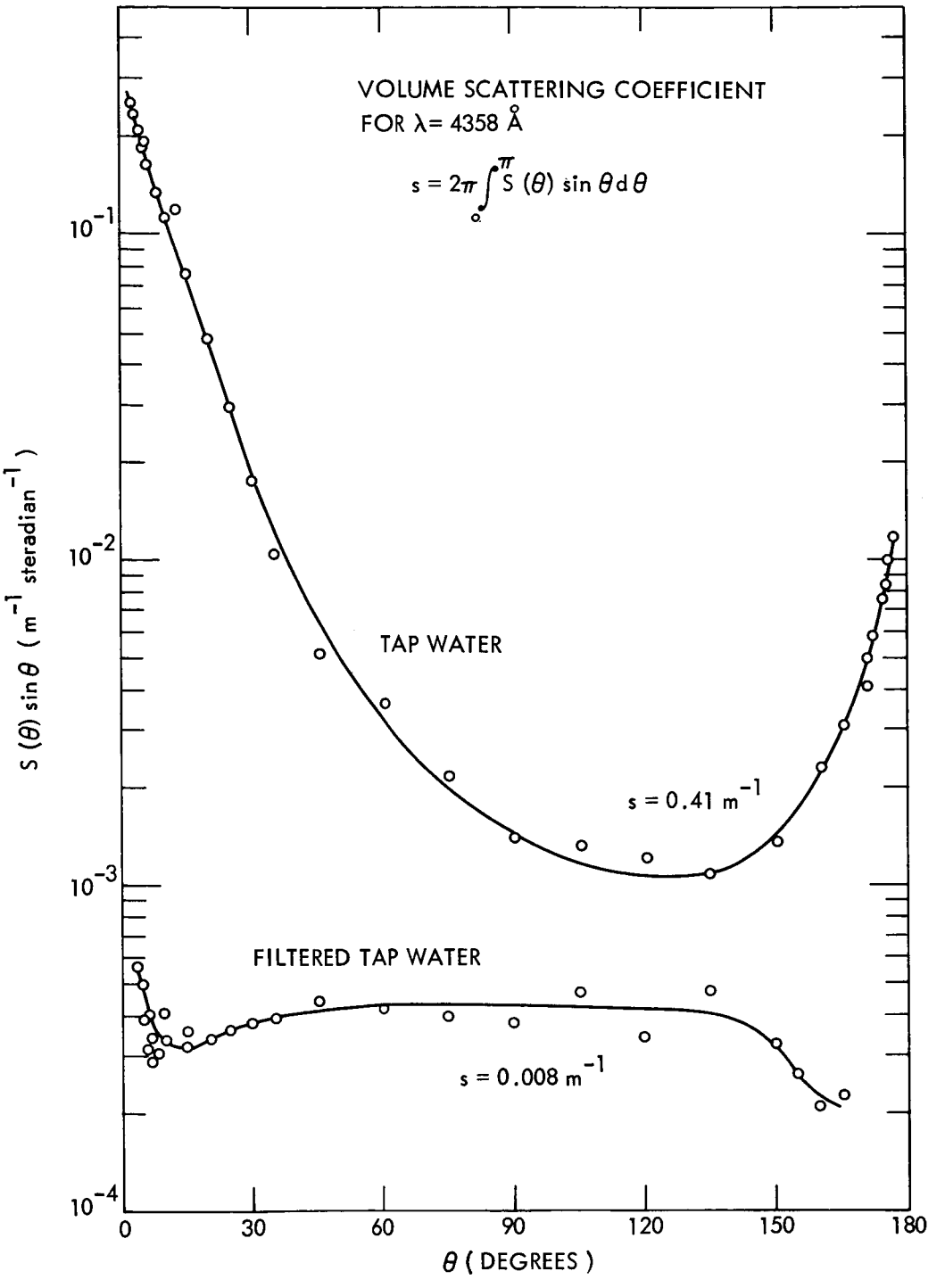
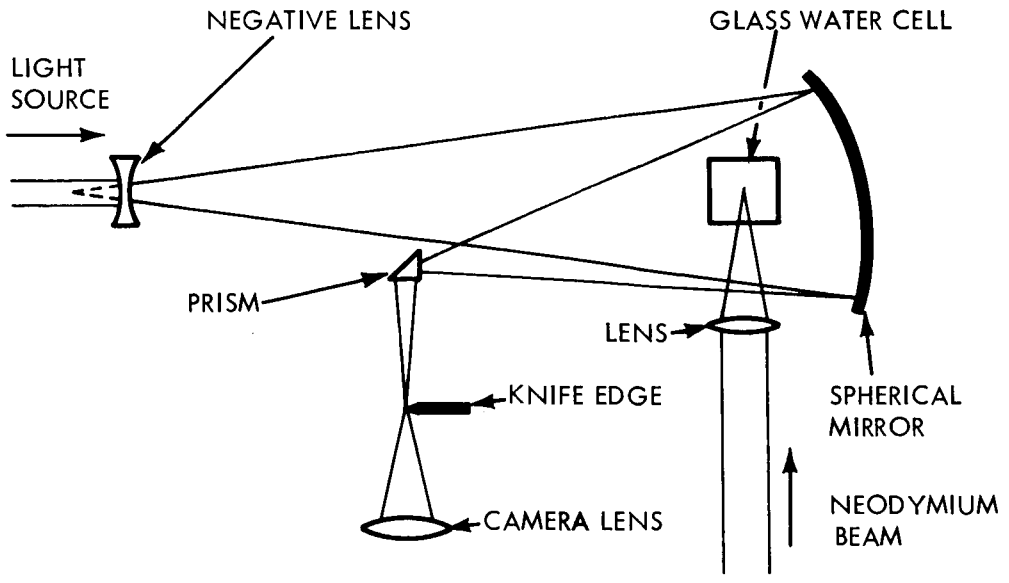
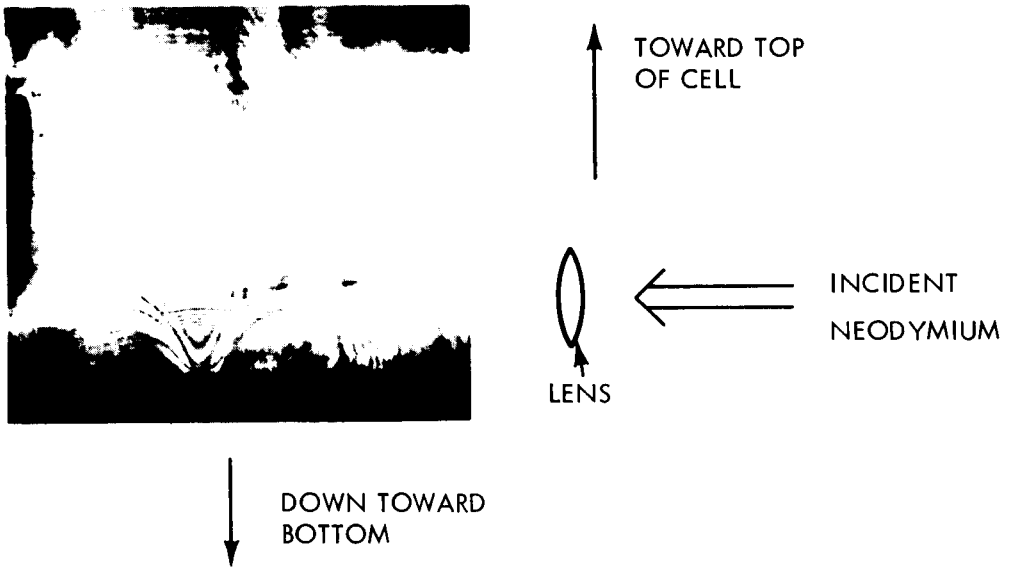


FIG. 10 VOLUME SCATTERING COEFFICIENT OF WATER





TYPICAL SCHLIEREN PHOTOGRAPH

FIG. 11 EXPERIMENTAL SCHLIEREN ARRANGEMENT

ENVIRONMENTAL MAGNETIC STUDIES

Two-Step Electron-Radiomagnetic Process Induces Extreme Rectangularity in Magnetic Hysteresis Loops of Nickel-Iron Alloys

D. I. Gordon
R. S. Sery

A one-step electron radiomagnetic treatment (electron irradiation in the presence of an applied magnetic field at 75 to 135° C) for inducing a high degree of uniaxial anisotropy and hence loop rectangularity in low coercive force nickel-iron alloys has been previously reported on. The two-step process consists of electron irradiation (~10^{17} 2-MeV e/cm^2) with or without an applied magnetic field at 50 to 95° C, followed by a low temperature (<200° C) anneal in the presence of an applied magnetic field. This new process produces similar properties but provides a greater degree of control over the end results, because the temperature can be controlled more accurately during the anneal in the furnace than during the electron irradiation. The irradiation is further simplified because the magnetic field need not be applied simultaneously with the irradiation. This latter advantage also permits radiomagnetic treatment of open magnetic circuit cores in addition to closed cores. Polycrystalline samples of Mo Permalloy and Mumetal given the two-step treatment begin to respond at ~100° C and 150° C respectively. Fig. 12 shows the effects on Mumetal of post-irradiation one-hour magnetic anneals at successively higher temperatures. The effect on unirradiated samples is shown for comparison. High rectangularity loop material finds extensive application in various non-linear magnetic devices.

Soft Magnetic Alloy Development

H. H. Helms, Jr.
J. G. Stewart
C. G. Reed

Investigation of ferrous magnetic alloys containing aluminum and/or silicon, together with special additives has continued. These alloys are of particular interest because of their stability in environments of radiation and stress, and because of their oxidation resistance. The ferrous alloys containing 8-10% silicon and 5-7% aluminum, commonly known as "Sendust", continue to create most interest because they have high initial permeabilities, good electrical resistivity, low magnetostriction and magnetocrystalline anisotropy, and excellent wear resistance.

Improvements in techniques for producing these alloys have been developed in order to overcome their poor room temperature ductility and to satisfy their critical alloy composition limits.

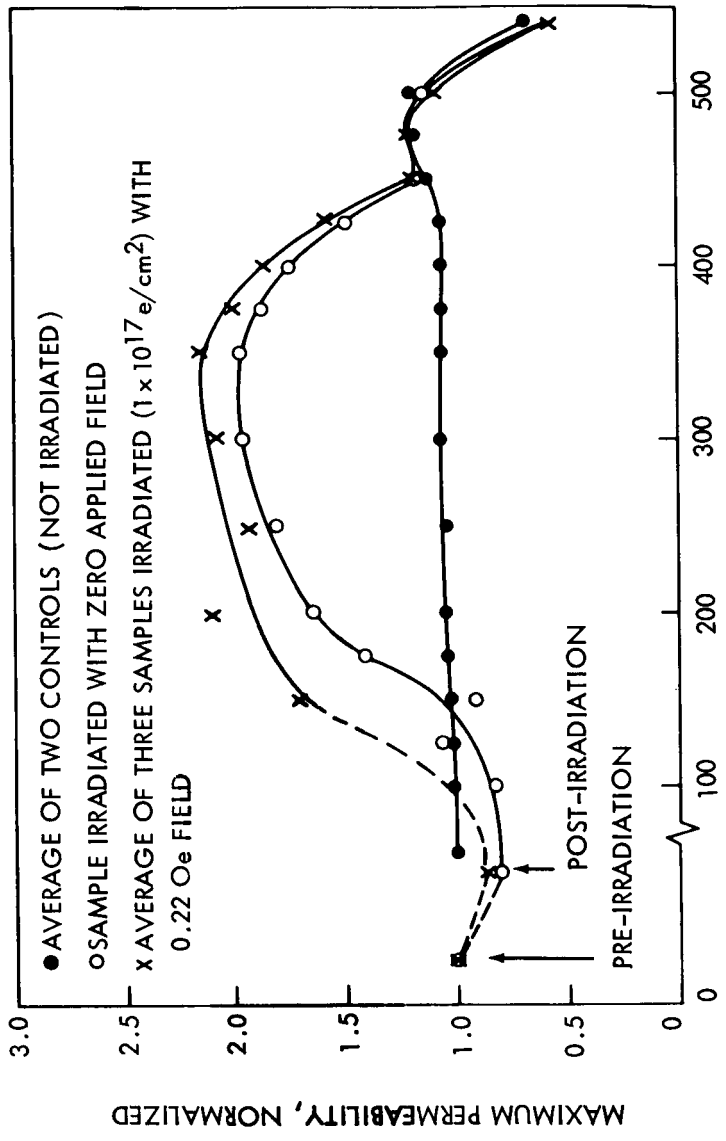


FIG. 12 EFFECTS OF 2-MeV ELECTRON IRRATIATION (WITH AND WITHOUT A FIELD) AND SUBSEQUENT ISOCHRONAL ANNEALING WITH A FIELD ON THE MAXIMUM PERMEABILITY OF MUMETAL

Hydrostatic compaction of Sendust and/or elemental powders in evacuated rubber containers at pressures up to 180,000 psi has been studied to observe the effects of this type of compaction vs the ordinary die compaction technique. Initial experiments have provided slight improvement in magnetic properties as a result of hydrostatic compaction of small samples. The dimensional changes during sintering were also easier to control than was the case with die compacted samples. The improved uniformity in density resulting from hydrostatic compaction is even more noticeable in preparation of thicker samples than in preparation of the samples having a smaller cross-section.

Small percentages of boron, germanium, and niobium have been added to the Sendust compositions, in addition to those elements such as tin, antimony, phosphorus, sulphur, etc., that were reported in the previous annual report. Determination of optimum powder mesh size has also been under study with some advancements being made in this direction. A maximum d.c. permeability of 101,000 with an initial permeability of 37,000 and a coercive force of 0.035 has been obtained on a sendust powder core having 1% in tin addition and using powders of optimum mesh size and proper heat treatment.

Many Sendust sample parts have been processed and ground to extremely close tolerances as requested by a number of commercial activities. These parts have been forwarded to the companies for evaluation in recording head applications.

Interest in high sensitivity magnetometry for future space and underwater applications has demanded improved materials for use as sensor elements. Modified molybdenum-permalloy type alloys have been heavily cold reduced and given various heat treatments in an effort to obtain desired properties. One particular alloy, 6% molybdenum - 81% nickel-iron, appears quite promising because of its low coercive force (.026) and its good rectangularity in thin gauge form.

High Pressure/High Temperature

D. W. Ernst
M. Pasnak
J. E. Tydings

A study of the effect of high pressure temperatures on the rare earth compounds Nd_6Mn_{23}, Sm_6Mn_{23}, Gd_6Mn_{23}, Ho_6Mn_{23}, Tb_6Mn_{23}, Dy_6Mn_{23} and Tm_6Mn_{23} was carried out. Initially, the compounds appeared to undergo a transformation from their face-centered cubic structure. While X-ray powder data indicated a single phase material, microprobe photographs revealed islands of rare earth element and manganese, indicating that the compounds had dissociated under the pressure and temperature treatment. The

Nd, Sm and Gd compounds readily decomposed at pressures of 40,000 atm and 1500°C, but it was necessary to go to temperatures of 1800°C in order to decompose the Ho, Tb, Dy and Tm compounds.

The R_6Mn_{23} compounds are size factor ones where the calculated A/B ratio is 1.31. High pressure by pushing the atoms closer together at high temperatures affected this ratio causing the compounds to decompose. The Nd, Sm and Gd decompose easier since they have a larger A/B ratio (1.33) and at the same time a lower melting point than the Ho, Tb, Dy and Tm compounds which have a more stable ratio (1.27) and a higher melting point.

MAGNETIC DEVICES

Ferromagnetic Microwave Device

E. T. Hooper, Jr.
A. D. Krall

Advancement was made in the development of the ferromagnetic microwave amplifier. Application of double pumping, where two distinct klystrons are used to pump the amplifier, was demonstrated. This technique resulted in an increased gain-bandwidth product greater than 20, along with a decrease in noise figure. The increased complexity of using two klystrons was offset by the discovery of psuedo-double pumping. With psuedo-double pumping the proper modulation on a single klystron accomplishes comparable results to double pumping. A simple procedure for aligning the amplifying crystal was devised. This procedure aligned the proper crystal orientation to reduce the anisotropy effects on resonance without the use of time consuming and costly X-ray techniques. A detailed investigation was made of the field configurations of the magnetostatic modes essential to the operation of the amplifiers. Evaluation of the results reveal that conventional techniques for excitation and suppression of modes are unworkable and must be modified in more sophisticated applications.

Gradiometer

E. T. Hooper, Jr.
W. M. Hubbard
J. F. Haben
J. Terrell

The proven techniques of ring-core magnetometery utilizing the sensor as the switching element in a saturable core-transistor multivibrator and cube-law detection have been extended into the field of gradiometry. A differential magnetometer with 10cm baseline has been developed with a sensitivity of 1 gamma/cm.

Thin 4% Mo - 79% Ni - 17% Fe tape material in toroidal core configuration is used as the sensing elements, self-driven in conjunction with two switching transistors. Individual magnetometer outputs consisting of asymmetric field dependent modulations are detected, filtered and substracted to yield a differential field indication. Amplification and frequency modulation follow producing an audio tone proportional to the differential magnetic field appearing across the two sensors.

Due to the simplicity of the circuitry, power drain is kept below 200 milliwatts delivered by two small batteries operating through two voltage regulator circuits as seen in the block diagram of Fig. 13.

A ferrous metal locator designated Locator EX-15 has been developed based on this principle for use in on-board search of watercraft for contraband weapons in the Vietnam area. In this ruggedized and waterproof configuration the sensors are mounted in line in a 7/8 inch tubular probe with all printed circuit board - solid state electronics and battery compartment sections in the probe handle. The resultant weight of the three-foot probe and headphone is 3 1/2 pounds.

This and similar developmental gradiometers have been operationally evaluated by the Navy Research and Development Unit-Vietnam. Production of the Locator EX-15 has been requested.

Magnetic Field Reduction in an Expandable Superconducting Enclosure

R. E. Brown

Experiments are under way on a novel procedure for obtaining a low magnetic field inside a superconducting hollow cylinder. A hollow cylinder made from lead foil is flattened at room temperature, as at A in Fig. 14, and then reopened at 4.2°K, as at B, when the lead is superconducting. It is found that the magnetic field is lower inside the opened cylinder than outside. The field along the axis of the cylinder should be lowered by a factor equal to the ratio of the cross-sectional area of the cylinder while flattened to the cross-sectional area after opening as a result of fluxoid conservation. We have obtained axial field reduction factors as low as 2×10^{-3} although 10×10^{-3} is the more usual result. The magnetic field remaining inside the cylinder is very stable because any change is opposed by a readjustment in the persistent currents. A low field, stable field region such as this offers promise as a test environment for evaluating the performance of sensitive magnetometers. Cascading this expandable superconducting enclosure operation is an attractive possibility. If opening a single flattened cylinder produces a field reduction factor F, then opening N

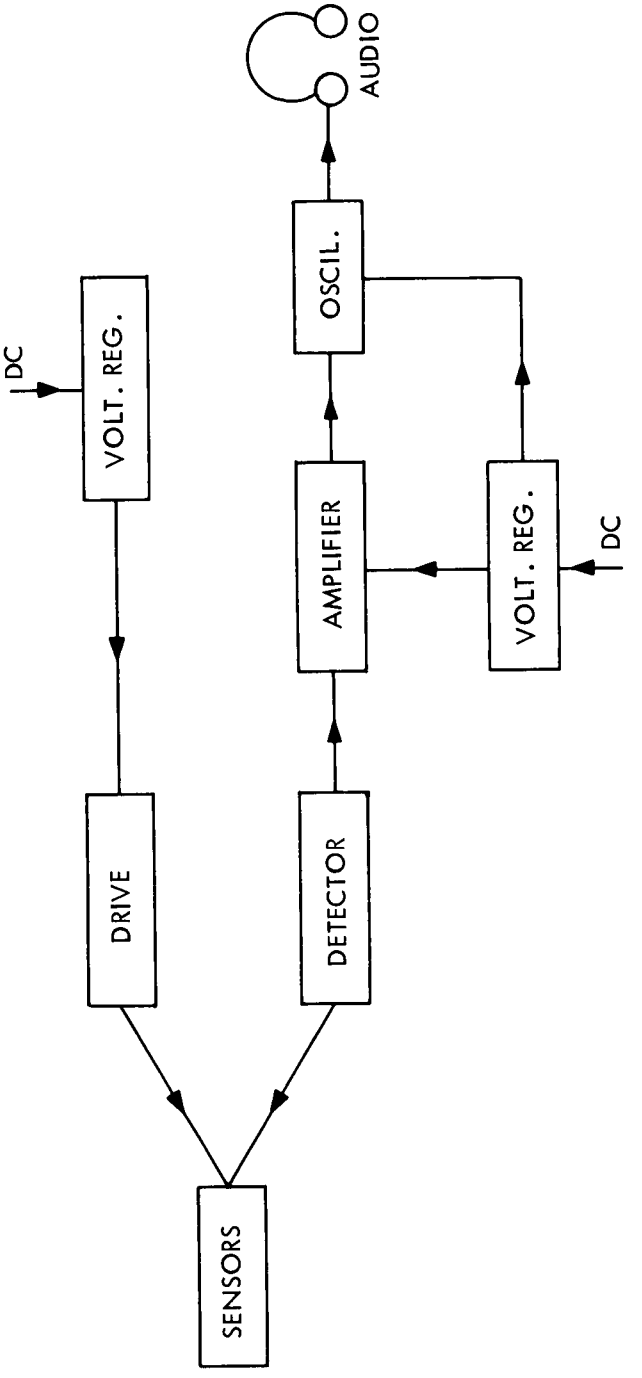


FIG. 13 BLOCK DIAGRAM OF LOCATOR EX-15

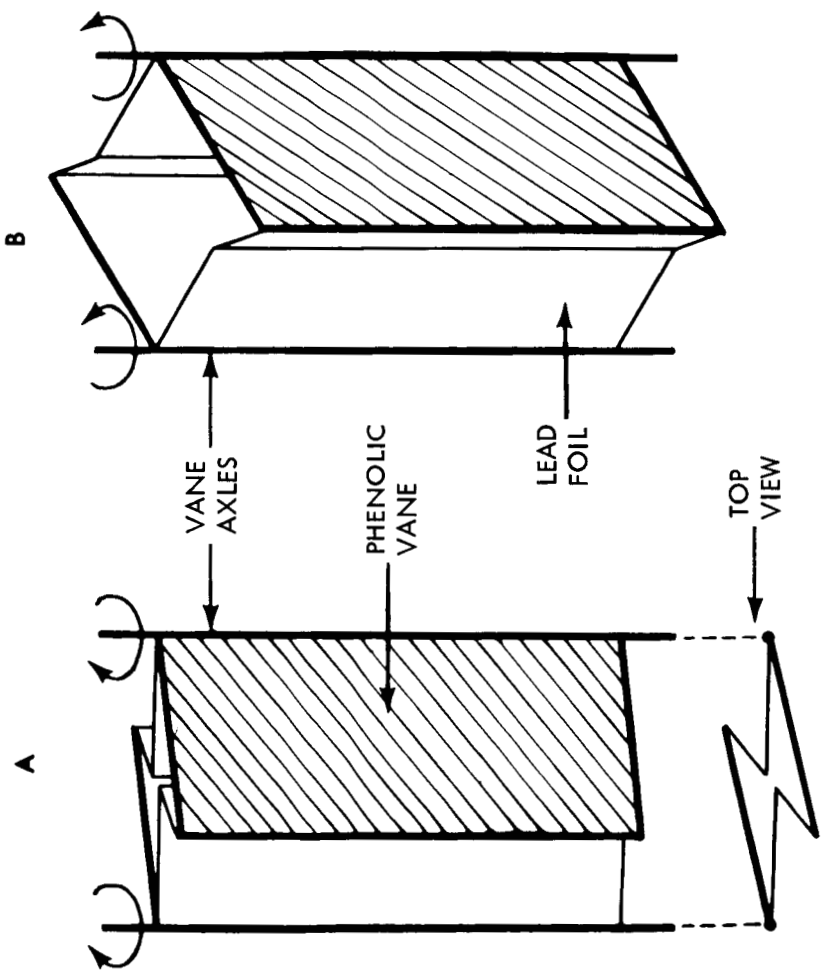


FIG. 14 SCHEMATIC DIAGRAM OF EXPANDABLE SUPERCONDUCTING ENCLOSURE

cylinders, one inside the other, should produce an over-all field reduction factor F^N. In principle the zero flux quantum state of a cylinder could be reached ultimately in which exactly zero magnetic flux links the cylinder axially.

We have carried this cascading procedure as far as the second stage. Starting at ambient earth's field reduction factors of 2×10^{-4} and 6×10^{-4} have been observed for the axial and transverse field components yielding a total field of about 10^{-4} oersteds near the center of the cylinders. Presumably the reduction factors will remain constant as the starting ambient field is decreased below earth's field since they depend in principle only upon the geometry of the device. In the easily attainable starting field of 10^{-3} oersteds total field levels approaching 10^{-7} oersteds may be achieved. Such fields are below the noise level of the best fluxgate magnetometers now being used in our measurements and will require improved techniques for detection. Future work will be directed toward increasing the number of cascaded stages and toward lowering the starting ambient field with a compensating coil system. Plans have been made for a scaled-up version providing a four-inch diameter room temperature access.

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The report summarizes the progress made in basic and applied solid state research in the Applied Physics Department of the U. S. Naval Ordnance Laboratory during 1965. The fields of research covered are metallic and non-metallic magnetic materials, semiconductors, infrared and metallurgy.

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